Aerosol-Cloud-Radiation Interactions in Atmospheric Forecast Models

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LONG-TERM GOALS

The long-term goal of this project is to gain a deep understanding of the role of atmospheric aerosols in affecting transmission of radiation through the atmosphere and in influencing cloud properties.

OBJECTIVES

The scientific objectives of this project are to identify the specific manner in which atmospheric aerosols determine cloud properties and to represent these interactions in atmospheric models. The technological objectives are to develop state-of-the-art instruments for aircraft sampling of aerosols that advance the long-term goals of the project.

APPROACH

The main technical approach is to conduct aircraft studies of the atmosphere, in which comprehensive sampling of atmospheric particles and radiative and cloud properties is carried out. The aircraft studies are complemented by laboratory investigations and theoretical analysis. Key individuals participating in this work are Professors John H. Seinfeld and Richard C. Flagan at the California Institute of Technology and Dr. Haf Jonsson at Naval Postgraduate School. Professor Seinfeld serves as Principal Investigator. Professor Flagan plays a key role in instrumentation development and planning of aircraft operations. As Chief Scientist of CIRPAS, Dr. Jonsson oversees all aspects of aircraft measurements and data management.

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14. ABSTRACT
The long-term goal of this project is to gain a deep understanding of the role of atmospheric aerosols in affecting transmission of radiation through the atmosphere and in influencing cloud properties.

15. SUBJECT TERMS

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WORK COMPLETED

During the past year, the work completed consists of the following:

1. Continued analysis of data from CSTRIPE field experiment (Monterey, CA, July 2003).

2. Analysis of data from ICARTT field experiment (Cleveland, OH, August 2004).

3. Conducting MASE field experiment (Monterey, CA, July 2005).

4. Development and application of models relating aerosols to cloud microphysical and radiative properties.

RESULTS

Tables 1 and 2 summarize the flights in the ICARTT and MASE field campaigns. Data from these campaigns are still being analyzed.

Table 1. ICARTT Twin Otter Flight and Instrument Performance Summary. Summary of the 12 flights flown in the ICARTT mission in Ohio, dated 8/2/04-8/21/04. Flight objectives included cloud profiling, and sampling of power plant plumes in cloudy and clear sky conditions. Flight coordination with the Meterological Service of Canada Convair aircraft was performed on six of the flights.

ICARTT Twin Otter Flight and Instrument Performance Summary Table **Last updated:** 5/5/05

General Flight Information					General Aerosol			Aerosol Chemistry			Soot			External Particle Sizers				CIRPAS Misc			
RF	Flight Date	Flight Time (UTC)	Mission Type	Convair Coord?	CCN	CVI	DMA	AMS	Filters	PILS	Photo-acoustic	PSAP	SP2	APS	CAS *	FSSP	PCASP	MET	NAV	CPC	Gerber LWC
1	8/2/04	15:07-20:32	Test / Aerosol Characterization	yes	down	N/A	down	OK	OK	OK	> 50%	down	OK	OK	> 50%	OK	OK	OK	> 50% (T)	OK	OK
2	8/3/04	16:57-21:52	Clouds S of Cleveland	no	> 50%	down	OK	OK	OK	OK	OK	down	OK	OK	> 50%	OK	OK	OK	> 50% (T)	OK	OK
3	8/6/04	16:17-20:41	Conesville PP plume/cloud	yes	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	> 50%	OK	OK	OK	> 50% (C)	OK	OK
4	8/8/04	18:18-21:45	Conesville PP plume/clear air	no	OK	N/A	OK	OK	OK	OK	OK	OK	OK	OK	> 50%	OK	OK	OK	> 50% (C)	OK	OK
5	8/9/04	17:09-22:16	Conesville PP plume/cloud	no	OK	down	OK	OK	OK	OK	OK	OK	OK	OK	> 50%	OK	OK	OK	> 50% (C)	OK	OK
6	8/10/04	18:04-23:00	Monroe PP plume/cloud	no	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	> 50%	OK	OK	OK	> 50% (C)	OK	OK
7	8/11/04	17:54-22:46	Cloud physics, SE shore of Erie	no	down	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	> 50% (C)	OK	OK
8	8/13/04	18:31-23:03	Detroit/Monroe PP plume	yes	OK	OK	OK	down	OK	OK	OK	OK	OK	OK	OK	down	OK	OK	> 50% (C)	OK	OK
9	8/16/04	18:16-22:37	Cloud physics, SW of Cleveland	yes	OK	OK	OK	N/A	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	> 50% (C)	OK	OK
10	8/17/04	18:13-21:24	Cloud physics, SW of Cleveland	yes	OK	OK	OK	> 50%	down	OK	OK	OK	OK	OK	OK	OK	OK	OK	> 50% (C)	OK	OK
11	8/18/04	15:37-19:10	Clouds, SW Ontario	yes	OK	OK	< 50%	> 50%	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	> 50% (C)	OK	OK
12	8/21/04	17:40-22:52	Conesville PP plume/cloud	no	N/A	OK	OK	down	OK	OK	OK	OK	OK	OK	OK	OK	OK	OK	> 50% (C)	OK	OK

(PP = power plant)

* CAS: >50% means up to a few percents of data not usable (saturated in cloud)

Instrument performance legend:

OK	Instrument was on and functioning well during the entire flight
> 50%	Instrument was functioning for most of the flight (suffered some loss of data quantity/quality)
< 50%	Instrument was functioning for some time during flight (suffered major loss of data quantity/quality)
down	Instrument was not functioning, or there were other errors resulting in no good data for this flight
N/A	Instrument was not on board or not turned on during flight

Note on navigation data:
instrument in parentheses was not functioning (T = TansVector, C = C-MIGITS) Some redundancy exists, so all parameters may still be available even if one system did not function

Table 2. MASE Twin Otter Flight and Instrument Performance Summary. Summary of the 13 flights flown in the MASE mission in Monterey, CA, dated 7/2/05-7/17/05. Flight objectives included sampling of ship tracks in cloudy and clear sky conditions, and unperturbed marine clouds. Flight coordination with the DOE G1 aircraft was performed on four flights. Five flights in the mission have been identified as suitable for detailed analysis of ship track observations.

MASE Twin Otter Flight and Instrument Performance Summary Table **Last updated:** 8/31/05

General Flight Information					Aerosol Physics and Chemistry				Soot		Optical Particle Counters					Radiometry		CIRPAS Misc		
RF	Flight Date	Flight Time (UTC)	Mission Type	Coord w/ G1?	CVI	DMA	AMS	PILS	Photo-acoustic	PSAP	CPC	PDI	PCASP	FSSP	CAS	STRAP	nadir radiom	MET	NAV	Gerber LWC
1	7/2/05	20:03-22:48	south, parallel to coast	-	OK	> 50%	OK	OK	OK	OK	OK	down	OK	OK	OK	OK?	OK	OK	OK	OK
2	7/3/05	17:02-21:18	south, perpendicular to coast	-	OK	OK	OK	OK	OK	OK	OK	< 50%	OK	OK	OK	> 50%	OK	OK	OK	OK
3	7/5/05	16:56-21:00	ship track	-	OK	OK	OK	OK	OK	OK	OK	> 50%	OK	OK	OK	OK	OK	OK	OK	OK
4	7/6/05	16:56-19:54	Pt. Reyes / G1 coord	yes	OK	OK	OK	OK	OK	OK	OK*	OK	OK	OK	OK	OK	OK	OK	OK	OK
5	7/8/05	16:59-21:01	front, line in ocean	-	OK	down	OK	OK	OK	OK	OK*	OK?	OK	OK	OK	> 50%	OK	OK	OK	OK
6	7/9/05	17:00-20:06	possible ship track	-	OK	OK	OK	OK	OK	OK	OK*	> 50%	OK	OK	OK	> 50%	OK	OK	OK	OK
7	7/10/05	17:00-21:18	clouds & clear air	-	OK	OK	OK	> 50%	OK	OK	> 50%	> 50%	OK	OK	OK	< 50%	OK	OK	OK	OK
8	7/11/05	18:57-20:55	SJV, Lenschow, OPC view volume char	-	N/A	OK	OK	OK	OK	OK	OK*	< 50%	OK	OK	OK	< 50%	OK	OK	OK	OK
9	7/13/05	17:18-20:50	ship tracks	-	OK	OK	OK	OK	OK	OK	OK*	OK	OK	OK	OK	OK	OK	OK	OK	OK
10	7/14/05	17:30-21:17	ship track	-	OK	OK	down	OK	OK	OK	OK*	OK	OK	OK	OK	OK	OK	OK	OK	OK
11	7/15/05	17:15-20:36	G1 coord, clean clouds, ship track, long run	yes	OK	OK	OK	OK	OK	OK	OK*	> 50%	OK	OK	OK	> 50%	OK	OK	OK	OK
12	7/16/05	17:23-21:50	G1 coord, CVI char, G1 exhaust, clear cond	yes	OK	OK	OK	OK	OK	OK	OK	> 50%	OK	OK	OK	> 50%	OK	OK	OK	OK
13	7/17/05	16:59-21:22	G1 coord, ship sighting & track	yes	OK	OK	OK	OK	OK	OK	OK*	> 50%	OK	OK	OK	> 50%	OK	OK	OK	OK
S1	7/4/05		Ferry to Sacramento MHR	-	N/A	N/A	OK	OK	> 50%	OK	OK	N/A	OK	OK	OK	N/A	N/A	OK	OK	OK
S2	7/4/05		Ferry from Sacramento MHR	-	N/A	OK	down	OK	OK	OK	OK	N/A	OK	OK	OK	N/A	N/A	OK	OK	OK

* CPC 3025 flow was low, but can be corrected by dividing N by 2.436; cutoff not 3 nm

"Golden day"
"Platinum day!"

Instrument performance legend:

OK	Instrument was on and functioning well during the entire flight
> 50%	Instrument was functioning for most of the flight (suffered some loss of data quantity/quality)
< 50%	Instrument was functioning for some time during flight (suffered major loss of data quantity/quality)
down	Instrument was not functioning, or there were other errors resulting in no good data for this flight
N/A	Instrument was not on board or not turned on during flight

CSTRIPE CCN Data Analysis

The California Institute of Technology's (Caltech) three-column cloud condensation nuclei (CCN) instrument (CCNC3) was deployed on the Center for Interdisciplinary Remotely Piloted Aircraft Studies (CIRPAS) Twin Otter as part of the Coastal Stratocumulus Imposed Perturbation Experiment (CSTRIPE) campaign that took place during July 2003 in Marina, CA. The goal of this study is to determine the extent to which we can model aerosol activation in actual clouds. The CCNC3 obtained CCN concentrations simultaneously at three different supersaturations (s) on 10 flights and at two different supersaturations on 7 flights during CSTRIPE. An aerosol/CCN closure study was undertaken using predictions from an activation model based on Köhler Theory. The model calculates the critical supersaturation for particles that contain certain soluble salts, certain organics, and generalized insoluble material. The observed CCN concentration (N_O) is then compared to the CCN concentration predicted (N_P) from the Köhler Theory model and measured aerosol size

distributions from the Caltech Dual Automatic Classified Aerosol Detector (DACAD). Preliminary closure analysis for all flights, assuming an aerosol composition of 100% ammonium sulfate ($(NH_4)_2SO_4$), results in mean (μ) closure ratios (N_P/N_O) of: $\mu(N_P/N_O)$=1.52 at s =0.09%, $\mu(N_P/N_O)$=1.95 at s =0.28%, $\mu(N_P/N_O)$=1.38 at s =0.58%. A $\mu(N_P/N_O)$ greater than unity indicates that fewer activated particles were observed than are predicted from Köhler Theory with particles composed of 100% $(NH_4)_2SO_4$ and the size distributions from the DACAD. This could result from insoluble or organic material within internally or externally mixed particles, which will be further studied by incorporating aerosol compositional data from the Aerodyne Aerosol Mass Spectrometer (AMS) into the Köhler Theory model. Four different atmospheric conditions were sampled during the CSTRIPE campaign– urban-influenced valley, fire-influenced continental, marine, and flare-perturbed marine. Differences in CCN properties are observed between these conditions, as well as between supersaturations within the same conditions. Future work will divide the flights into separate air masses, which will allow further investigation of CCN properties of different aerosols that were sampled during the same flight. Compositional data from the Caltech AMS and assumed insoluble volume fractions and external mixing properties will be included in the calculation of N_P in closure analyses to further understand the CCN properties of the sampled aerosol.

Marine Stratus Experiment (MASE)

MASE provided an opportunity to study "ship tracks" as a semi-controlled laboratory for aerosol-cloud interactions. Of the 13 TO marine flights that were conducted during MASE, six encountered strong, localized perturbations in aerosol concentration, size and composition measurements consistent with ship emissions. These emissions and their impact on the stratus layer were analyzed using the detailed cloud profiling strategy that has been successful in previous missions. Strong effects of the enhanced aerosol loading were found in cloud droplet concentration and droplet size distributions. Cloud albedo generally had a maximum value over a ship track, although natural variability in cloud albedo associated with turbulent cloud structure was large compared to the expected signature due to aerosols. Results from the flight on July 5 are highlighted here (Figure 1). Two neighboring ship tracks were studied on this flight. The chemistry measurements, comprising the Aerodyne TOF-AMS (flown for the first time during MASE), the PILS, and the photoacoustic absorption device (from which black carbon concentration is inferred), showed a number of relevant features: 1) a fairly concentrated layer of organic carbon aerosol (~3 $\mu g\ m^{-3}$), probably of urban origin, overlay the marine boundary layer; 2) Within the MBL, organic aerosol loading was lower (~0.5 $\mu g\ m^{-3}$), however, sulfate aerosol loading was comparable to that overlying the MBL (~ 1 $\mu g\ m^{-3}$); 3) the shiptracks are associated with a very strong enhancement in MBL sulfate aerosol (~ 0.5 to 2 $\mu g\ m^{-3}$) above the background, but virtually no observable enhancement in organic or black carbon aerosol was found.

The aerosol and cloud data look promising to address the following questions:

1) Does the increase in droplet concentration correspond to an increase or decrease in droplet dispersion?

2) Is precipitation different in the track compared with outside of the track?

3) Is there a substantial difference in turbulent dynamics, humidity, liquid water content, or heat content within a track compared to the background?

4) Is cloud base systematically different within a track?

5) Is the enhancement of cloud albedo over tracks statistically significant, when interpreted with the aid of the cloud microphysical measurements?

6) Can questions not answerable for individual tracks be answered by a statistical analysis of the ensemble of tracks studied during MASE?

7) Are the relationships seen in the data consistent with those predicted by detailed 3-D RAMS simulations?;

8) is the cloud response to day-to-day variations in the background aerosol concentration similar to that to the ship-track forcings, or are there systematic differences linked to differences in the characteristics of the aerosol perturbation and the time-scales of the cloud response?

9) To what degree is the organic aerosol layer overriding the cloud gradually “seeding” the MBL with CCN, and is this layer an effective mechanism for isolating aerosol from the wet removal process and increasing the influence of the indirect effect in remote regions?

Preliminary data analysis suggests that at least 3 of the other tracks have a data quality comparable to that seen on July 5.

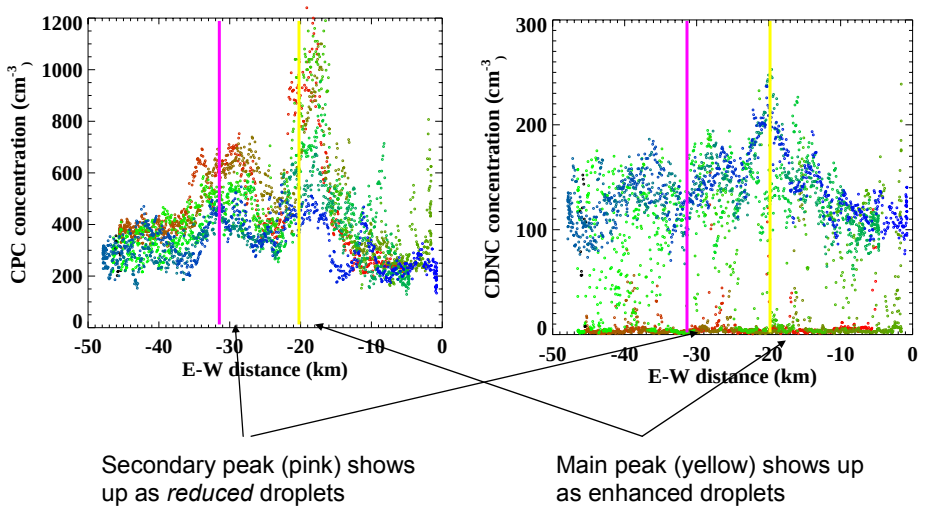

Figure 1: Characteristics of Ship Track sampled during MASE on July 5, 2005. Preliminary modeling suggests that the differences in the aerosol size distributions are largely responsible for the strong differences in the cloud's microphysical responses to the aerosol perturbations.

Particle-into-Liquid (PILS) Sampler

The particle-into-liquid sampler (PILS), developed under an ONR DURIP grant, quantifies the chemical composition of ambient particles. The PILS samples sub-micron particles and grows them into droplets sufficiently large to be collected by inertial impaction. The droplets are deposited into vials held on a rotating carousel. The liquid sample in each vial can be partitioned and analyzed by different techniques, mainly ion chromatography (IC). The PILS and IC cooperatively can determine the ambient air concentration of water-soluble species, specifically inorganic ions and organic acids. Figures 2 and 3 show PILS data from the August 2004 ICARTT campaign.

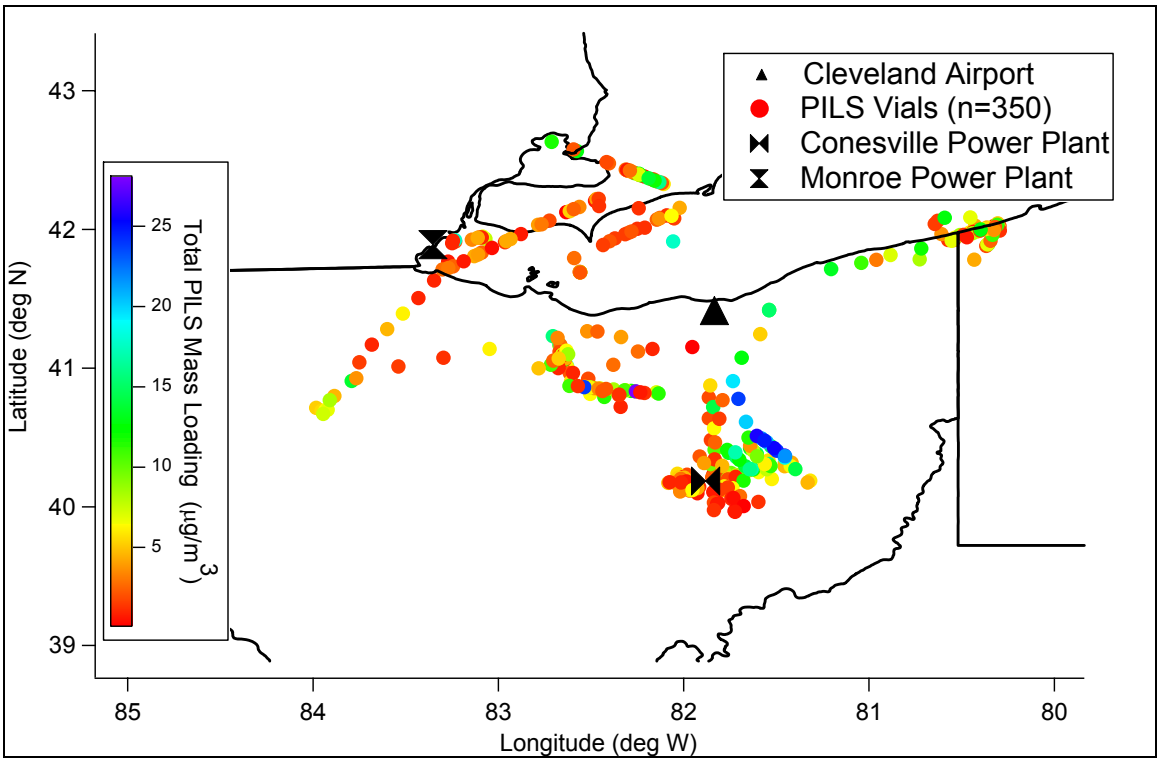

Figure 2. Sampling region of the Twin Otter during ICARTT with markers indicating PILS vials that were collected and the colors representing the magnitude of the total sub-micron aerosol mass loading measured.

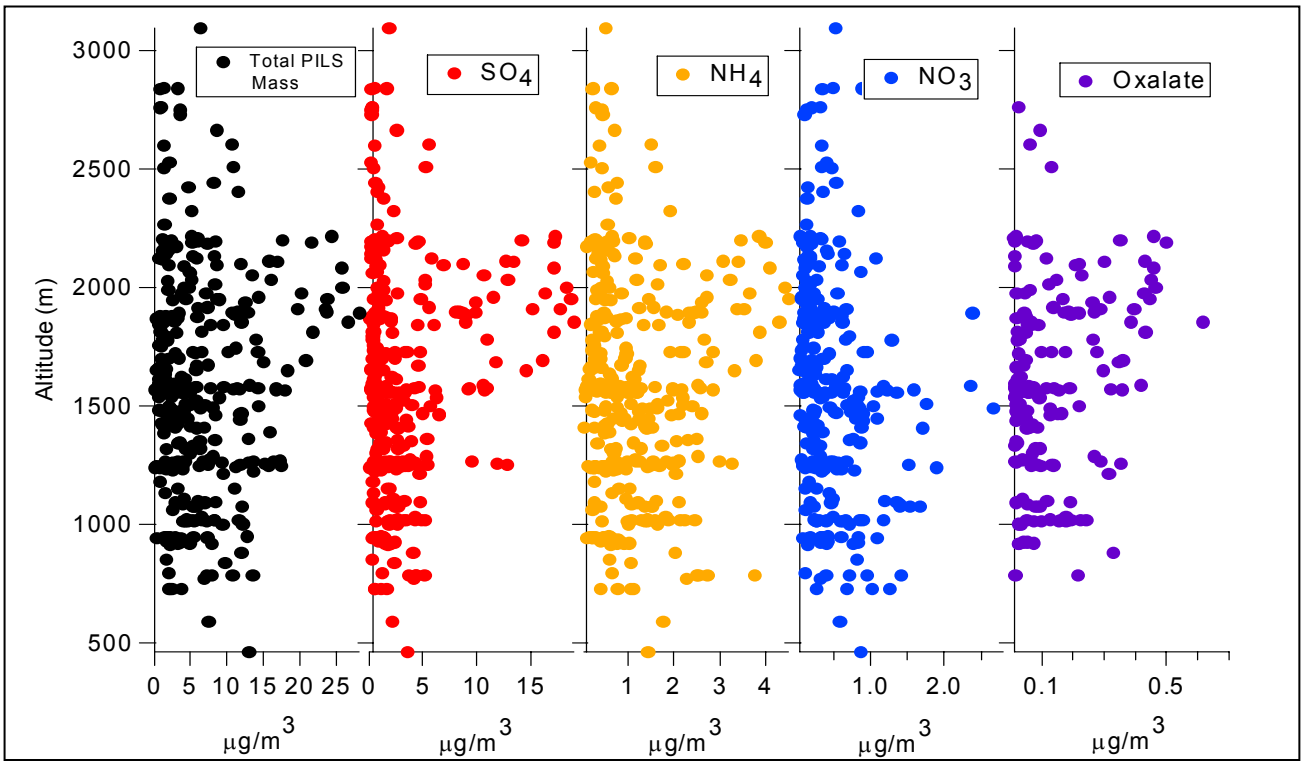

Figure 3. Vertical distributions of specific ions measured by the PILS during ICARTT. The Twin Otter was mainly sampling below 2.5 km. The measured PILS mass concentrations start to decrease close to 3 km indicating that the Twin Otter was occasionally sampling close to the dividing point between the free troposphere and the mixed layer below it. The PILS measured the highest mass loadings downwind of the Conesville Power Plant, a coal-burning power generation facility, with the maximum being approximately 28 µg/m3. Sulfate dominated the sub-micron particulate ionic mass, and this is most likely from secondary formation from SO2. Ammonium was the next biggest contributor to the ionic mass and it was highly correlated with sulfate. Nitrate usually stayed below 1 µg/m3, and dropped to its lowest levels when the PILS was sampling under acidic conditions downwind of power plants. Significant levels of oxalate were measured in cloudy conditions indicating that aqueous phase chemistry is integral to the production of this dicarboxylic acid.

Variations in the Cloud Liquid Water Path as a Result of the Increase in Aerosol Number Concentrations

We utilized the LES RAMS model (Large Eddy Simulation Regional Atmospheric Model System) coupled with the explicit bin-resolved cloud microphysics model (LES RAMS-bin model) [*Tzivion, et al.*, 1987, 1989; *Feingold, et al.*, 1994; *Stevens, et al.*, 1996; *Stevens, et al.*, 1998 ; and successfully investigated cloud liquid water variations due to aerosol number concentration changes [*Lu and Seinfeld,* 2005a]. In this theoretical study we selected two well-studied marine stratocumulus cases: The first one is the sounding profile based on the FIRE [First ISCCP (International Satellite Cloud Climatology Project) Regional Experiment] from *Moeng et al.* [1996], representing a weakly drizzling marine stratocumulus; while the second sounding profile from *Stevens, et al.* [1998], based on the ASTEX (Atlantic Stratocumulus Transition Experiment), represents a strongly drizzling marine stratocumulus nocturnal case. The overall work is based on a series of 98 three-dimensional LES simulations of marine stratocumulus clouds under both nighttime and daytime conditions, and a wide range of aerosol number concentration spanning from clean to polluted (N_a = 50–2500 cm^{-3}) and the different co-varying meteorological conditions (sea surface temperature (SST), large-scale divergence, two sounding profiles). Through the statistical correlations, we have found that τ (cloud optical depth) is both positively correlated with N_a and LWP, with a higher correlation of τ with LWP than it with N_a. Moreover, we showed that the two dynamical factors (SST and divergence) may exert an effect on cloud optical depth as large as or even greater than that exerted by varying microphysical properties (N_a). Additional simulation results of the giant sea salt CCNs show that they have negligible effect on τ for the FIRE case, but they result in a reduction on τ of 3%–77% for polluted ASTEX clouds (N_a = 1000–2500 cm^{-3}). Therefore, the simulation results suggest the impact of giant sea salt is more important for moist and potentially convective clouds.

Variations in the Cloud Spectral Dispersion as a Result of the Increase in Aerosol Number Concentrations

We further explored the factors that control the cloud spectral relative dispersion d (ratio of cloud droplet spectral width to the mean radius of the distribution) as a result of aerosol number concentration changes [*Lu and Seinfeld,* 2005b]. Results show that an enhancement of the cloud susceptibility (the change of cloud optical depth due to change of cloud droplet number concentration) results from the positive dependence of the coefficient k (relating cloud droplet effective radius and volume mean radius in the large-scale models) on the aerosol number concentration. This positive correlation of k with N_a is mainly due to the inverse relationship of d with N_a. We found that the

decreasing of d with increasing N_a (for $N_a \lesssim 1000$ cm^{-3}) are because smaller droplets resulting from higher aerosol number concentrations inhibit precipitation and lead to these physical mechanisms:

(1) less spectral broadening by suppressed collision and coalescence processes;

(2) more spectral narrowing by droplet condensational growth at higher updraft velocity, because reduced drizzle latent heating at cloud top results in increased boundary layer turbulent kinetic energy production by buoyancy and thereby stronger turbulence.

Increased spectral broadening owing to increased cloud-top entrainment mixing, also as a result of increased boundary layer turbulence, is relatively insignificant compared with (1) and (2). Simulation results also suggest that neglect of spectral skewness and drizzle drops as typically in calculating k [e.g., *Pontikis and Hicks*, 1992; *Martin, et al.*, 1994] overestimates k, and it will therefore, underestimates the dispersion effect on cloud susceptibility, especially for strongly drizzling clouds. In summary, the maximum enhancements of cloud susceptibility as a result of the cloud spectral dispersion effect alone by about 4.2% and 39% for simulated FIRE and ASTEX cases, respectively.

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